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Amorphous Silicon Film Deposition by Low Temperature Catalytic Chemical Vapor Deposition (<150 °C) and Laser Crystallization for Polycrystalline Silicon Thin-Film Transistor Application

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We deposited amorphous silicon (a-Si) films below 150 °C with a custom-designed catalytic chemical vapor deposition (Cat-CVD) system. The hydrogen content of the films was controlled at less than 1.5 at. %. Excimer laser crystallization was performed without the preliminary dehydrogenation process. Crystallization occurred at a laser energy density above 70 mJ/cm². Thin-film transistors (TFTs) were fabricated while the entire process temperatures were maintained between 200 °C. We obtained a field-effect mobility of higher than 100 cm²/V s and a sub-threshold slope of 116 mV/dec. The a-Si film prepared by a low temperature Cat-CVD is a promising candidate for polycrystalline silicon TFTs of the active matrix display.

[keywords: catalytic CVD (Cat-CVD), low temperature deposition, amorphous silicon (a-Si), hydrogen content (C₃H), excimer laser annealing (ELA), polycrystalline silicon (poly-Si), thin film transistor (TFT)]

Recently, low temperature polycrystalline silicon (LTPS) thin-film transistors (TFTs) has attracted much attention for active matrix flat panel displays (AMFPDs) or flexible display because LTPS TFTs have greater mobility compared to amorphous silicon (a-Si) TFTs.¹⁻³ The key process of LTPS TFT is crystallizing the silicon films. Excimer laser annealing (ELA) is an effective method for obtaining the improved channel by crystallizing the a-Si. However, if the deposited a-Si films have a large content of hydrogen (C₃H), bubbling or ablation may occur during the laser crystallization. In general, a-Si films prepared by plasma enhanced chemical vapor deposition (PECVD) method have a C₃H of approximately 10 at. %. Therefore, a heat treatment step at approximately 500 °C is needed to minimize the C₃H before crystallization, but such high temperature may also impose another problem, especially when plastic substrates are used for flexible display panels.

Catalytic chemical vapor deposition (Cat-CVD) is expected to be one of the promising methods for obtaining high quality a-Si layers with low C₃H. It has several advantages compared to PECVD. Especially, a-Si films prepared by Cat-CVD contain atomic hydrogen below 3 at. %,⁴⁻⁵ This level of C₃H is much lower than that obtained by the conventional PECVD method, and the subsequent crystallization can be performed without the prior thermal process for dehydrogenation.⁶⁻⁷

In conventional Cat-CVD, the process temperature increases to ~300 °C during the deposition. This temperature necessitates the use of quartz or glass substrate. However, the substrate temperature should be kept below 200 °C for the plastic application. Figure 1 shows the scheme of the Cat-CVD apparatus used in this experiment. To decrease the substrate temperature, we adopted a custom-designed cooling system around the chamber, catalyzer, and substrate holder. We deposit a-Si on various substrates during one minute, and confirmed that the substrate temperature was maintained at below the glass-transition temperature of the plastic substrate.⁷⁻⁸

Two main deposition parameters, chamber pressure and source gas flow rate, were optimized to produce the lowest possible hydrogen content (C₃H). Other deposition parameters are listed in Table I. The C₃H of as-deposited a-Si films was estimated by Fourier-transformed infrared spectroscopy (FTIR).⁸⁻⁹

a-Si films of 50 nm thick were deposited on a corning 1737 glass with SiO₂ buffer layer of 2000 Å so as to minimize the laser damage. The deposited buffer layer should suppress a diffusion of impurities from the glass substrate into a-Si films during the laser crystallization as well.¹⁰ The film thickness of a-Si was controlled by adjusting the process time. A high deposition rate of 120 nm/min was obtained.

We used a XeCl excimer laser with a wave length of

Table I. Deposition condition.

<table>
<thead>
<tr>
<th>Substrate temperature (°C)</th>
<th>150</th>
</tr>
</thead>
<tbody>
<tr>
<td>W wire temperature (°C)</td>
<td>1700–1800</td>
</tr>
<tr>
<td>W wire-substrate distance (cm)</td>
<td>3</td>
</tr>
<tr>
<td>SiH₄ gas flow (sccm)</td>
<td>1</td>
</tr>
<tr>
<td>Deposition pressure (mTorr)</td>
<td>2</td>
</tr>
</tbody>
</table>

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308 nm and a pulse duration of 30 ns, for crystallization of a-Si films. Laser energy density was selected from 60 to 180 mJ/cm². We used a multi-step laser irradiation method in order to prevent the sudden evolution of hydrogen.\(^{11}\) The laser energy density was increased by 10 mJ/cm² for each step, and a single pulse per step was utilized. The crystallinity of the a-Si films were evaluated by using an optical microscope and UV-reflectance spectroscopy.\(^{12,13}\) The grain size was measured by using a scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Prior to the SEM analysis, the Secco etching was performed to enhance the visibility of the grain boundary. Top-gate n-type TFTs were fabricated using the Cat-CVD a-Si precursor films. The channel width was 20 \(\mu\)m and the length was 8 \(\mu\)m. All process temperatures were kept below 200 °C so as to apply to the plastic substrate in the future. We also used multi-step laser irradiation method with a final laser energy density of 130 mJ/cm². A glass buffer layer and gate insulator was deposited using inductively coupled plasma (ICP)-CVD of high density plasma at 170 °C,\(^{14}\) and gate metal was deposited using an E-beam evaporator. Activation annealing was performed using a self-aligned ELA after the gate dopant ion implantation.

During the deposition process, the heat from the catalyzer is transferred to the substrate mainly by radiation. Therefore, effective blocking of the radiation is a key factor to decrease the substrate temperature. We successfully suppressed the substrate temperature below 150 °C by adding the cooling system for a period of 1 min. Figure 1 shows the Cat-CVD and cooling system used. The cooling water around the filament and around the chamber wall can minimize the re-radiation from the heated chamber wall by the catalyzer. The substrate can be cooled by circulating water through into the sample holder.\(^{7}\) In general, the thickness of the silicon films required for TFT active layer is 400 –1000 \(\AA\). The Cat-CVD has a deposition rate that is higher than 120 nm/min. Therefore, the deposition process can be finished within only 30 sec, and the substrate temperature can be kept even below 100 °C during deposition.

The hydrogen content (C\(_{\text{H}}\)) of a-Si is the most important factor for laser crystallization. In the Cat-CVD, the chamber pressure and source gas flow rate are the main parameters that determine the deposition rate and the quality of a-Si films. The C\(_{\text{H}}\) also largely depends on these parameters. Figure 2 shows the variation of C\(_{\text{H}}\) with the process parameters. C\(_{\text{H}}\) decreased as either the chamber pressure or the source gas flow rate decreased. These results agree well with the work of Jadkar et al.\(^{15,16}\) At a lower pressure and a lower gas flow rate, a C\(_{\text{H}}\) of less than 1.5 at. % was obtained. This level is sufficiently low to perform laser crystallization without the dehydrogenation step. In the Cat-CVD, the density of decomposed species is higher than that of the PECVD. Basically, the Cat-CVD has a high deposition rate. In addition to it, a high density of H atoms in the gas phase may remove other H atoms from the growing surface of films. Consequently, the amount of residual H atoms inside the films becomes much lower than those in the PECVD.

Direct ELA was successfully carried out without the preliminary dehydrogenation step. The multi-step laser irradiation method was used to minimize damage due to abrupt effusion of hydrogen. The interval between the steps is 10 mJ/cm² and the laser irradiated sequentially from 60 to 180 mJ/cm². In Fig. 3(a) of UV-reflectance spectroscopy; peaks at the wavelength of 280 nm and at 370 nm indicate the formation of the crystalline phase. The degree of crystallization can be calculated from the peak intensity.\(^{12,13}\) The line with circular symbols represents the a-Si film before ELA, and those with triangular and diamond symbols represent laser-annealed films, respectively. As the crystalline fraction in the film is larger, the peak intensity becomes strong. The crystallization was started at 70 mJ/cm², and the films were almost fully crystallized when the laser energy density reached to 90 mJ/cm².

Figure 3(b) summarizes the degree of crystallinity of the silicon films. The crystallinity saturates to almost 90%, at above 90 mJ/cm². Correspondingly, the grain size was estimated by the SEM as shown Fig. 4(a).

In Fig. 4(a), we can observe conspicuous grains. The average grain size was rather small, approximately 50 nm.
The grain size was not drastically dependent on the final laser energy density. Figure 4(b) shows the cross-sectional TEM image. We observed the columnar structure of polycrystalline silicon (poly-Si) layer which is peculiar to the deposition by the low pressure chemical vapor deposition (LP-CVD) at temperatures higher than 600 °C. We also confirm that the Si surface region shows a rather high crystallinity. It is speculated that the initial amorphous phase from the Si films deposited by the Cat-CVD is different from what was obtained by conventional PECVD. As only a small amount of hydrogen is contained in the films, the Si films can endure a rather high energy of laser irradiation. After pulsed laser irradiation, the columnar structure could be formed initially as the temperature in the film increases. The vertical structure limits subsequent lateral grain growth. More work needs to be done to clarify the detailed growth mechanism.

Figure 5 shows the $I_d-V_g$ characteristics of the top-gate n-type TFT fabricated with the Cat-CVD silicon films. We obtained a field-effect mobility value higher than 100 cm$^2$/V·s and a steep sub-threshold slope of 116 mV. Even though the average grain size was small, the peak mobility was rather high. This result suggests that although our polycrystalline films have small grains, surface morphology should be rather smooth. For the top-gate type TFT, the channel appears initially near the surface of poly-Si layer. The surface smoothness is important. As shown in the TEM pictures in Fig. 4, there exists a narrow poly-Si region of good crystallinity at the interface with the gate dielectric layer, and this region may provide a good path for electron transport, resulting in good field-effect mobility values.

Another thing to be noted is that the field-effect mobility value decreased drastically with increases in the gate voltage. The TFT characteristic is considered to be related to the columnar grain structure, and a detailed relationship must be clarified further. The TFT having this level of mobility values is expected to drive the organic light emitting diodes (OLEDs) if an Cat-CVD apparatus for large area substrates is developed.